Automated Advanced Calibration and Optimisation of Thermochemical Models Applied to Biomass Gasification and Pyrolysis

N. Bianco[1], M. C. Paul[2], G. P. E. Brownbridge[1]*, D. Nurkowski[1], A. M. Salem[2], U. Kumar[2], A. N. Bhave[1], M. Kraft[3]

[1] Computational Modelling Cambridge Ltd. (CMCL Innovations), Cambridge - UK

[2] Systems, Power and Energy Research Division, School of Engineering, University of Glasgow, Glasgow - UK

[3] Department of Chemical Engineering and Biotechnology, University of Cambridge - UK

** Corresponding author: Dr George P. E. Brownbridge, gbrownbridge@cmclinnovations.com*

ABSTRACT

This article aims to combine physico-chemical modelling to statistical analysis algorithms to provide alternative advanced approach for the optimisation of biomass pyrolysis and gasification processes. The goal was to develop an automated flexible approach for the analyses and optimisation of these processes. The approach can also be directly extended to other biomass conversion processes, and in general to all those processes for which a parameterised model is available. A flexible physico-chemical model of the process is initially built. Within this model, a hierarchy of sensitive model parameters and input variables are identified, which are then automatically adjusted to calibrate the model and optimise the process. Through the mathematical model of the process we can understand how species concentration and reactor conditions evolve in the system under study. The flexibility given by the ability to control any parameter of the model is critical in providing the capacity to effectively control both the efficiency of the process and its emissions. It allows users to design and operate feedstock-flexible pyrolysis and gasification processes, accurately control product characteristics, and limit or prevent the formation of unwanted by-products (e.g. tar in biomass gasification processes).

INTRODUCTION

An alternative and sustainable way of producing energy, fuels and chemicals with low, zero or negative emissions and meeting the total demand globally is crucial for providing the security of future supply long after the conventional fossil fuels run out. Biomass gasification

and pyrolysis can ensure this due to feedstock sustainability, inherent carbon neutrality and ability to potentially achieve negative emissions when coupled with carbon capture and sequestration (CCS) technologies [1].

A large number of biomass feedstocks, including energy crops, wastes and wood pellets, can be transformed into energy, biofuels and other chemicals via two main conversion routes, thermochemical and biological. The thermochemical route is more flexible in terms of the feedstocks that can be used and, when compared to the biological route, its products are also more compatible with those of existing petroleum refining operations (transport fuels and feedstock chemicals) [2]. Thermochemical conversion processes include direct combustion, pyrolysis and gasification. As mentioned earlier, this article will focus on two of these processes, namely pyrolysis and gasification.

During pyrolysis, the biomass is thermally degraded by heat in the absence of oxygen at lower temperatures compared to those used for direct combustion and gasification. From the degradation of the biomass, charcoal, bio-oil (or pyrolysis oil) and fuel gas are produced. Bio-oil can be used as fuel in boilers, diesel engines or gas turbines [3].

Gasification occurs when partial oxidation of biomass at elevated temperatures is carried out. Oxidation reactions takes place in the biomass gasifier at temperatures between 700-1400°C. The primary product in this case is a mixture of gases, also known as syngas or producer gas, primarily composed of H_2, CO, CO_2 and CH_4. Syngas can be converted into fuels, including H_2, Fischer-Tropsch diesels and synthetic gasoline, as well as widely used chemicals such as methanol and urea [2].

To promote these processes, novel, cost-effective and more efficient technologies are required. One of the main challenges in biomass conversion processes like gasification and pyrolysis is to understand how operating conditions and feedstock composition affect the reactions within the process and, therefore, the product specifications [2]. Some of the main difficulties encountered when trying to model these correlations include the fact that biomass feedstock is often neither completely known nor homogeneous [4], the intrinsic reaction pathway of the pyrolysis reactions, central to the overall biomass thermal treatment process, are complex and not fully understood, and multiscale phenomena take place within the reactor [5].

In optimally exploiting the flexibility and supply security offered by various biomass feedstocks and the intrinsic advantages in terms of the low CO_2 footprint of biomass gasification and pyrolysis technologies, virtual engineering has a vital role to play as a key

enabler in accelerated cost-effective technology development and TRL progression [6] [7] [8] [9] [10]. The use of a flexible and reliable physico-chemical model, and the ability to speed up and automate its analysis, understanding, calibration and exploitation via integration with a statistical analysis toolkit, which is done by this work, will provide an effective and robust strategy to tackle and solve the drawbacks listed above. Through optimal design and detailed process control will allow us to achieve the feedstock flexibility, and product yield and quality, required to make biomass pyrolysis and gasification widely exploited processes.

Considerable effort has been spent in formulating suitable models for the intrisically complex biomass pyrolysis and gasification processes and in identifying optimal design, operating and kinetic parameters for these models. Kinetic and process parameters have been identified either through experimental [11] [12] [13] or numerical [14] [15] methods. Much less has been done in the field of statistical analysis, automated calibration and optimisation of these models [16] [17].

The availability of a large array of mathematical models is advantageous. Statistical analysis toolkits can be used to carry out rapid assessment of the model performances before any type of analysys or perameter estimation is implemented. A proprietary statistical analysis toolkit from CMCL Innovations, MoDS (Model Development Suite), is used here to perform global sensitivity analysis, error propagation studies, calibration and optimisation of available models for biomass pyrolysis and gasification processes, with MoDS able to seamlessly couple with these, as well as with virtually any other third party model or toolkit.

A novel strategy that automates the estimation and optimisation of model parameters and reaction rates is presented. Two proprietary software from CMCL Innovations, namely *k*inetics and MoDS, together with a thermochemical model for downdraft gasifiers developed by the research group of Dr Manosh Paul at the University of Glasgow, have been used to produce the results presented in this paper.

MODEL DESCRIPTION AND INTEGRATION WITH MoDS

MoDS is a statistical analysis software that finds use in a number of applications, including parameter estimation [18] [19] [20] [21], optimal design of experiment, global sensitivity and uncertainty propagation analysis [22] [23], and process optimisation. MoDS uses a suite of numerical and statistical analysis algorithms to gain insight from other models, simulators or CAE (Computer Aided Engineering) toolkits, treating them as black boxes. Results from these analysis can be used by the software to identify trends and provide predictions based on the original model [24]. MoDS was used here to calibrate available models for biomass

gasification and pyrolysis against existing experimental data, and to optimise a biomass downdraft gasification process.

Extensible Markup Language (XML) or comma delimited (CSV) files containing model parameters and control variables are automatically created by MODS and passed as inputs to the model, either through an API (Application Programming Interface) or as the input files to an executable generated from the model script. These files are generated iteratively by a sampling algorithms, such as Monte Carlo or Sobol [25] [26] [27] [28]. Results are produced generally in CSV format, and then read and analysed by MoDS, which uses the information gathered through this iterative process to perform, for example, global sensitivity analysis, optimal design of experiment, model calibration against experimental data or process optimisation.

Biomass pyrolysis

*k*inetics is a software used in the automotive/non-road, energy, and chemical processing industries to build, manage and implement chemical kinetic models. It comprises a library of chemical reactor (physico-chemical) models derived from first principles [29] [30]. Examples of applications of *k*inetics, and of the *k*inetics-based suite for IC (Internal Combustion) engine and fuel modelling kinetics & SRM Engine Suite, include the development of reduced chemical kinetic schemes for fuel oxidation and emission pathways [31] [32], and the modelling of organic [33] [34] and inorganic [35] nanoparticles synthesis.

The analysis of chemical kinetics can become challenging as the number of interacting chemical species increases. A sensitivity analysis is required in this cases, with *k*inetics that can be used to perform local sensitivity analysis, as well as mechanism reduction on any of its models.

The biomass pyrolysis kinetic mechanism used here was taken from Ranzi et al. [36]. It defines the biomass as a mixture of three key components: cellulose, hemicellulose and lignin. These components can be mixed in different proportions depending on the type of biomass (e.g. softwood, hardwood, etc.). The model provides a lumped kinetic scheme describing the devolatilisation of these components, and the decomposition of the solid into permanent gases, condensable vapours (tars) and solid residues (char). The detailed kinetic scheme is completed by the mechanism for the secondary reactions of the released gas-phase species. A constant pressure homogeneous batch reactor was used to simulate the biomass pyrolysis process in *k*inetics.

The coupling between the model developed in *k*inetics and MoDS is realised through a workflow. The MoDS-*k*inetics workflow was designed to provide a seamless handover between these two software. The reactor model setup and process conditions are defined in *k*inetics. These are automatically exported in MoDS, where advanced statistical analysis and calibration of the model is performed. A post-processing toolkit (CMCL Explorer) has been also integrated within the workflow and is used to represent the dynamic evolution of both model inputs and calibrated parameters, as well as results from the sensitivity analysis and error propagation studies performed on the model. A schematic of the workflow used for model calibration is presented in Figure 1.

Any model parameter available to *k*inetics can be estimated using MoDS via the workflow. MoDS provides recommended calibration settings and can validate settings that the user customises. Experimental data can be read directly from comma delimited files. MoDS automatically interpolates profile data to compare model and experiments at different times or reactor lengths [37]. The optimisation algorithm choice and configuration can be tailored to fit the specific model and problem.

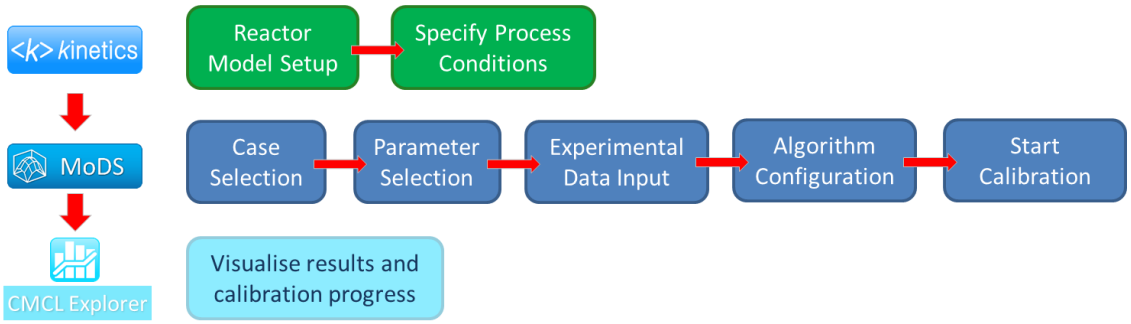


Figure 1 MoDS-kinetics workflow for model calibration

Biomass gasification

The model used here is a four-zone (namely drying, pyrolysis, oxidation and reduction zones) integrated 1D kinetic model of a biomass downdraft gasifier, which allows to investigate the effect of, for example, moisture content and air-to-fuel ratio on the operating conditions and design specifications of each zone, as well as to predict composition and tar content of the producer gas. Individual chemical kinetic schemes are used for each zone of the gasifier to describe the main mechanisms of biomass drying, pyrolysis, oxidation and reduction [38] [39].

The model is written in Matlab. A wrapper for the Matlab code was created to generate a model executable with input and output files, which was used by MoDS. The MoDS-Matlab

integrated toolkit was used to perform an initial analysis and calibration of the model, and subsequently to optimise the downdraft biomass gasification process with the aim of identifying design specifications and operating conditions for the gasifier leading to a reduction in tar and other undesired gaseous species, and to an increase in the heating value and yield of the syngas.

RESULTS AND DISCUSSION

Biomass pyrolysis

Thermogravimetric (TG) data were taken from the heated wire mesh reactor studies of Milosavljevic and Suuberg [40], who studied cellulose pyrolysis at different heating rates. Simulations were conducted in *k*inetics using a constant pressure homogeneous batch reactor model with an imposed temperature profile. It was assumed that at the beginning of the process only cellulose was present (Y_{cell} = 1.0). Figure 2 shows the simulation results when a heating rate of 5 K/min is used. It can be seen that a good agreement with the experimental data is obtained, when combining the detailed mechanism from Ranzi et al. [41] [42] and the model developed in *k*inetics.

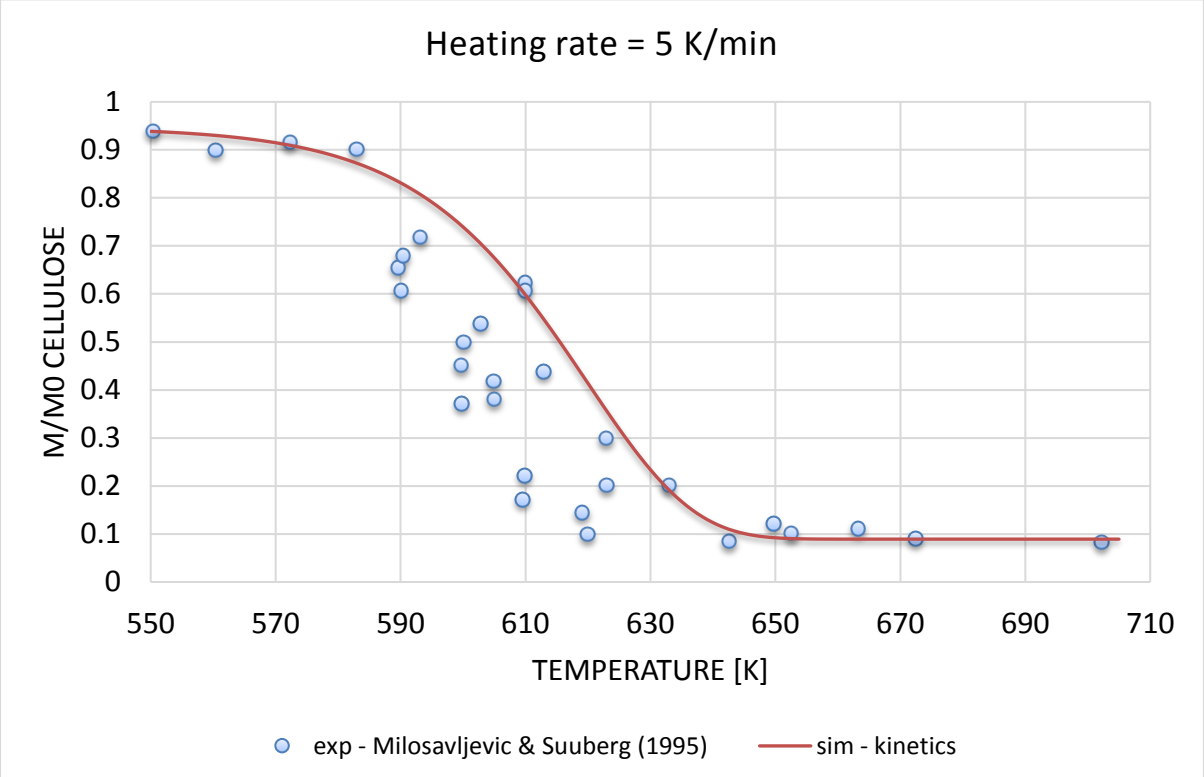


Figure 2 Non-isothermal thermogravimetric curves for the pyrolysis of cellulose in inert gas - model predictions (line) and experimental data (points) [40].

With the aim of further improving the match with the experimental data, we identified key reactions by performing a mechanism reduction. Available mechanism reduction strategies in *k*inetics were used to produce a hierarchy of reduced models. One of the algorithms used is the Directed Relation Graph with Error Propagation (DRGEP) [43]. Each node in a DRGEP represents a species in the detailed mechanism, while the edges and their thickness represent the importance of these species relatively to initially selected target species, which are those species we want to keep in the mechanism. By using the DRGEP technique, we were able to identify, after combining the results from different possible scenario and by selecting an adequate error threshold, out of more than 20,000 reactions of the original mechanism [41] [42], 19 key reactions. By using the MoDS-*k*inetics workflow to calibrate the frequency factor of these 19 reactions we were able to further improve the match with the experimental data, as can be seen in Figure 3. The original and modified values of these 19 frequency factor are reported in Table 1.

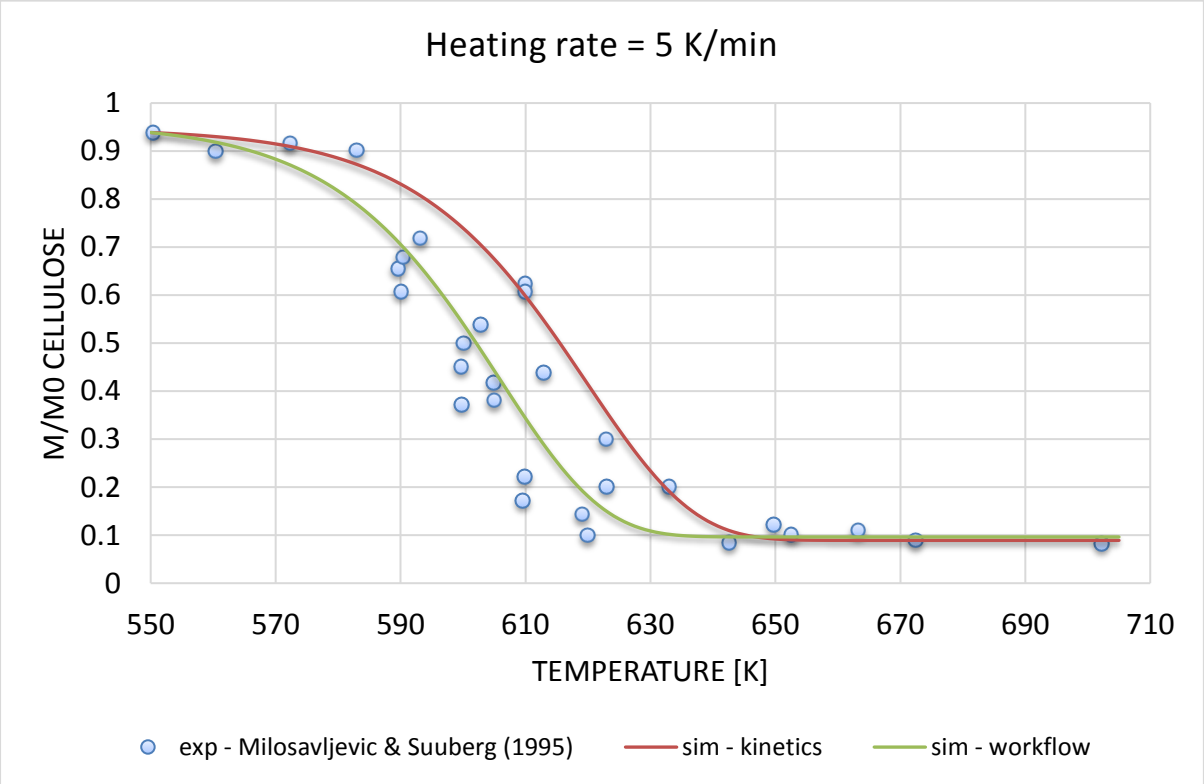


Figure 3 Non-isothermal thermogravimetric curves for the pyrolysis of cellulose in inert gas – original model (sim – kinetics) versus calibrated model (sim – workflow) predictions (points represent experimental data [40])

Levoglucosan (LVG) and hydroxyacetaldehyde (HAA) are between the major products of cellulose pyrolysis. Piskorz et al. [44] measured yields of those species produced during

pyrolysis of a microcrystalline cellulose (Avicel pH-102) at different temperatures and at an average residence time in the reactor of 0.5 seconds. The moisture content was set to Y_{H2O} = 2.9% [44]. The simulation time was set equal to the average experimental residence time. Obtained simulation data, when using the original model and the calibrated model with the frequency factors from Table 1, are presented in Figure 4. It can be seen that an improved agreement with the experimental data was achieved when using the calibrated model.

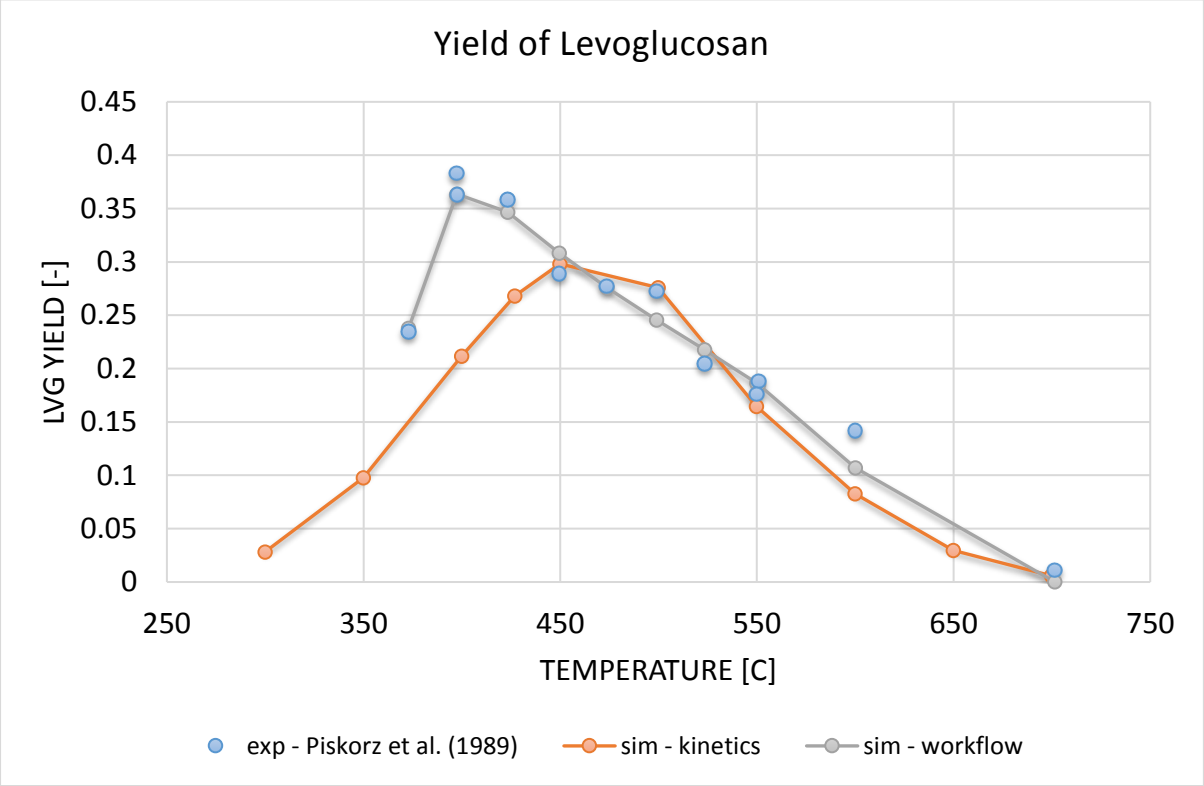


Figure 4 Yield of Levoglucosan from the pyrolysis of microcrystalline cellulose (Avicel pH-102) – original model (sim – kinetics) versus calibrated model (sim – workflow) predictions (points represent experimental data [44])

Similar results were also observed when implementing the workflow to more complex systems. Garcìa-Pérez et al. [45] produced TG curves for softwood bark residue and hardwood rich in fibres at different heating rates under nitrogen. Softwood and hardwood can be described as a combination of cellulose, hemicellulose and lignins of specified composition [46]. Following this strategy, and using the van Krevelen diagram to compute the feedstock composition from its elemental analysis [41], we were able to simulate the thermal decomposition of softwood and hardwood. Some of these results are presented, for softwood, in Figure 5. In the case presented here, a heating rate of 60 K/min was imposed. The original model is able to reproduce well the experimental results, but also in this case an

improved match is observed when using the modified 19 frequency factors of the calibrated model.

Table 1 Key reactions from the mechanism of Ranzi et al. [41] [42] with original and calibrated frequency factors

Reaction	**Original Frequency Factor**	**Calibrated Frequency Factor**	**Unit of measure**
CELLA → 5E-2*H_2 + 0.93*H_2O + 0.61*CO + 0.36*CO_2 + 0.3*CH_2O + 2E-2*HCOOH + 0.15*CH_3OH + 5E-2*$C_2H_2O_2$ + 0.15*CH_3CHO + 0.4*C_2H_4O2 + 0.35*C_2H_5CHO + 5E-2*$C_3H_6O_2$ + 0.25*$C_6H_6O_3$ + 0.61*CSOLID + 0.2*H_2(S) + 5E-2CH_4(S)	2.50E+06	7.91E+02	1/s
CELLA → $C_6H_{10}O_5$	3.30E+00	1.04E+03	1/K/s
CELL → 5*H_2O + 6*CSOLID	6.00E+07	6.00E+08	1/s
HCE1 → 0.4*H_2O + 0.16*CO + 0.12*$C_2H_2O_2$ + 0.2*$C_3H_6O_2$ + 0.2*$C_5H_4O_2$ + 0.6*$C_5H_8O_4$ + 8E-2*H_2(S)	3.00E+00	3.00E+02	1/K/s
HCE1→ 0.4*H_2O + 0.69*CO + 0.79*CO_2 + 0.3*CH_2O + 5E-2*HCOOH + 0.875*CSOLID + 1E-2*CO_2(S) + 1E-2*CO(S) + 0.9*COH_2(S) + 0.35*H_2(S) + 0.625*CH_4(S) + 0.375*C_2H_4(S)	1.80E-03	1.80E-06	1/K/s
HCE2 → CSOLID + 0.2*H_2O + 0.275*CO + 0.275*CO_2 + 0.4*CH_2O + 2.5E-2*HCOOH + 5E-2*$C_2H_4O_2$ + 0.35*CH_3COOH + 0.1*C_2H_5OH + 0.3*CO_2(S) + 0.725*COH_2(S) + 0.25*CH_4(S) + 0.3*CH_3OH(S) + 0.225*C_2H_4(S)	5.00E+09	1.58E+10	1/s
LIGC → H_2O + 0.32*CO + 0.3*CH_2O + 0.41*C_2H_4 + 8E-2*C_6H_5OH + 0.1*$C_9H_{10}O_2$ + 5.735*CSOLID + 0.35*LIGCC + 0.7*COH_2(S) + 0.495*CH_4(S)	1.00E+11	1.00E+14	1/s
LIGCC → H_2 + 0.7*H_2O + 1.4*CO + 0.65*CH_4 + 0.6*C_2H_4 + 0.35*$C_2H_4O_2$ + 0.2*C_6H_5OH + 0.3*$C_9H_{10}O_2$ + 6.75*CSOLID + 0.4*CO(S)	1.00E+04	1.00E+06	1/s
LIGOH → H_2O + 0.65*CO + 5E-2*CO_2 + 5E-2*HCOOH + 0.1*CH_4 + 0.6*CH_3OH + 0.1*C_2H_3CHO + 2.5E-2*$C_{24}H_{28}O_4$ + 4.25*CSOLID + 0.9*LIG + 0.6*CO(S) + 0.85*COH_2(S) + 5E-2*H_2(S) + 0.35*CH_4(S) + 0.3*CH_3OH(S) + 0.2*C2H4(S)	1.00E+08	1.00E+07	1/s
LIG → 0.3*CO + 0.3*CH_3CHO + 0.3*$C_6H_5OCH_3$ + 0.7*$C_{11}H_{12}O_4$ + 0.3*CO(S)	4.00E+00	4.00E+01	1/K/s
LIG → 6*CSOLID + 2*COH_2(S) + 0.6*H_2O + 0.4*CO + 0.4*CH_2O + 0.2*CH_4 + 0.2*CO(S) + 0.4*CH_4(S) + 0.4*CH_3OH(S) + 0.5*C_2H_4(S)	8.30E-02	8.30E-06	1/K/s
LIG → C_2H_4 + 0.6*H_2O + 2.6*CO + 0.4*CH_2O + 1.1*CH_4 + 0.4*CH_3OH + 4.5*CSOLID	1.00E+07	1.00E+03	1/s
CO_2(S) → CO_2	1.00E+06	3.16E+06	1/s
CO(S) → CO	5.00E+12	5.00E+14	1/s
COH_2(S)→ H_2 + CO	1.50E+12	1.50E+14	1/s
H_2(S) → H_2	5.00E+11	5.00E+13	1/s
CH_4(S) → CH_4	5.00E+12	5.00E+15	1/s
CH_3OH(S) → CH_3OH	2.00E+12	6.32E+14	1/s
C_2H_4(S) → C_2H_4	5.00E+12	5.00E+15	1/s

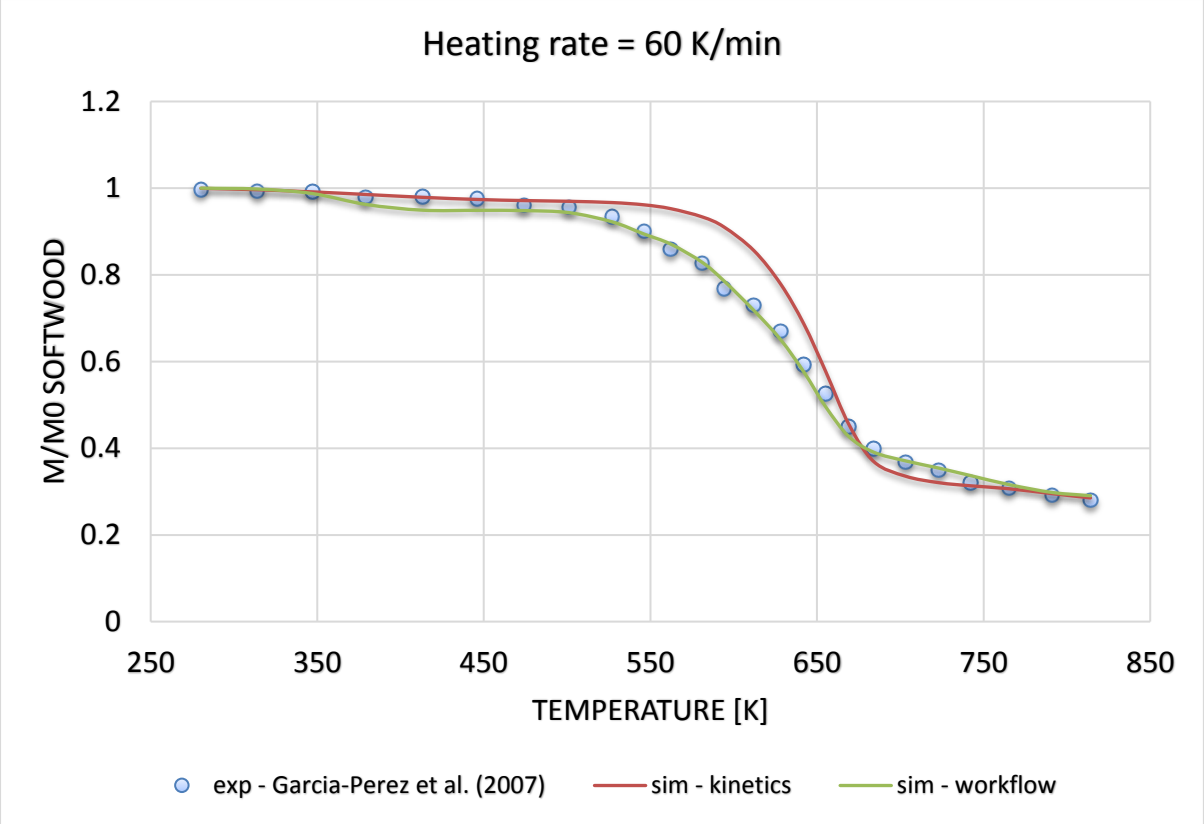


Figure 5 Non-isothermal thermogravimetric curves for the pyrolysis of softwood in nitrogen – original model (sim – kinetics) versus calibrated model (sim – workflow) predictions (points represent experimental data [45])

Biomass gasification

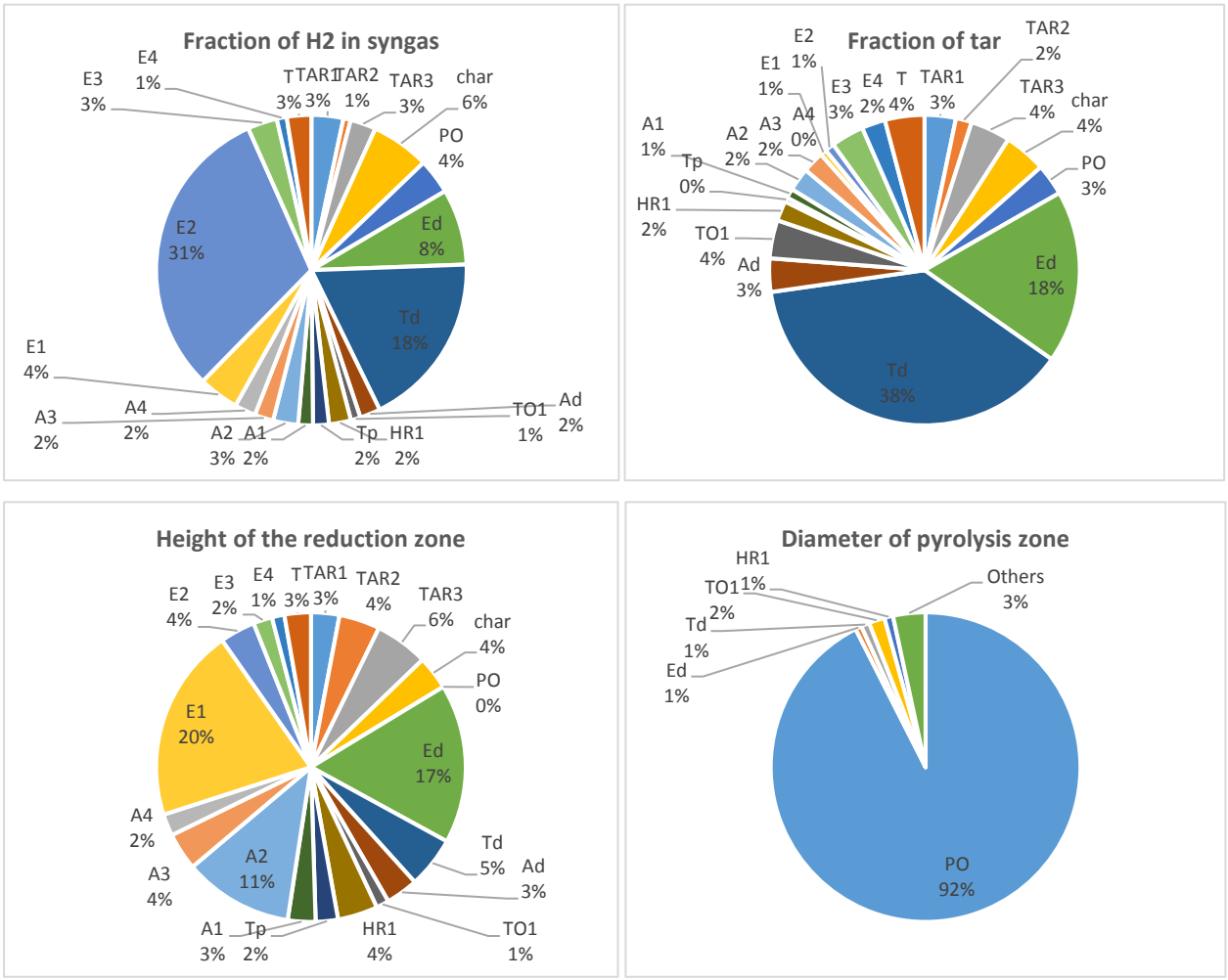
An initial study was carried out on a 20 kW biomass downdraft gasifier model from Salem and Paul [38], a 1D computationally non-intensive robust and flexible thermochemical model validated against a number of experimental results for different feedstocks. Sensitivity analysis and uncertainty propagation studies were performed on the model using MoDS. Some of the results from this study are presented in Figure 6 and Figure 7. Acronyms used in these figures are listed and explained in Table 2. Sensitivity analysis and uncertainty propagation studies were produced through MoDS for all the process outputs and design parameters of the model [38]. Only a selected limited number of results are presented here.

The study was performed by initially generating an HDMR (High Dimensional Model Representation) of the model. An HDMR is a multidimensional response surface, produced by MoDS as surrogate of the model. The global sensitivity of each model parameter, or combination of interconnected parameters (see Figure 7), are proportional to, and can be directly inferred from, the coefficients of the response surface (overall contribution to variance).

The results presented in Figure 6 and Figure 7 were produced using Neem, with a 20% moisture content, as feedstock [47]. A representative number of other samples were studied here, including bamboo, rubber wood, wood pellets, wheat straw and wood chips. The results from the studies carried out on all these samples were combined to identify the more sensitive inputs and uncertain outputs in the model.

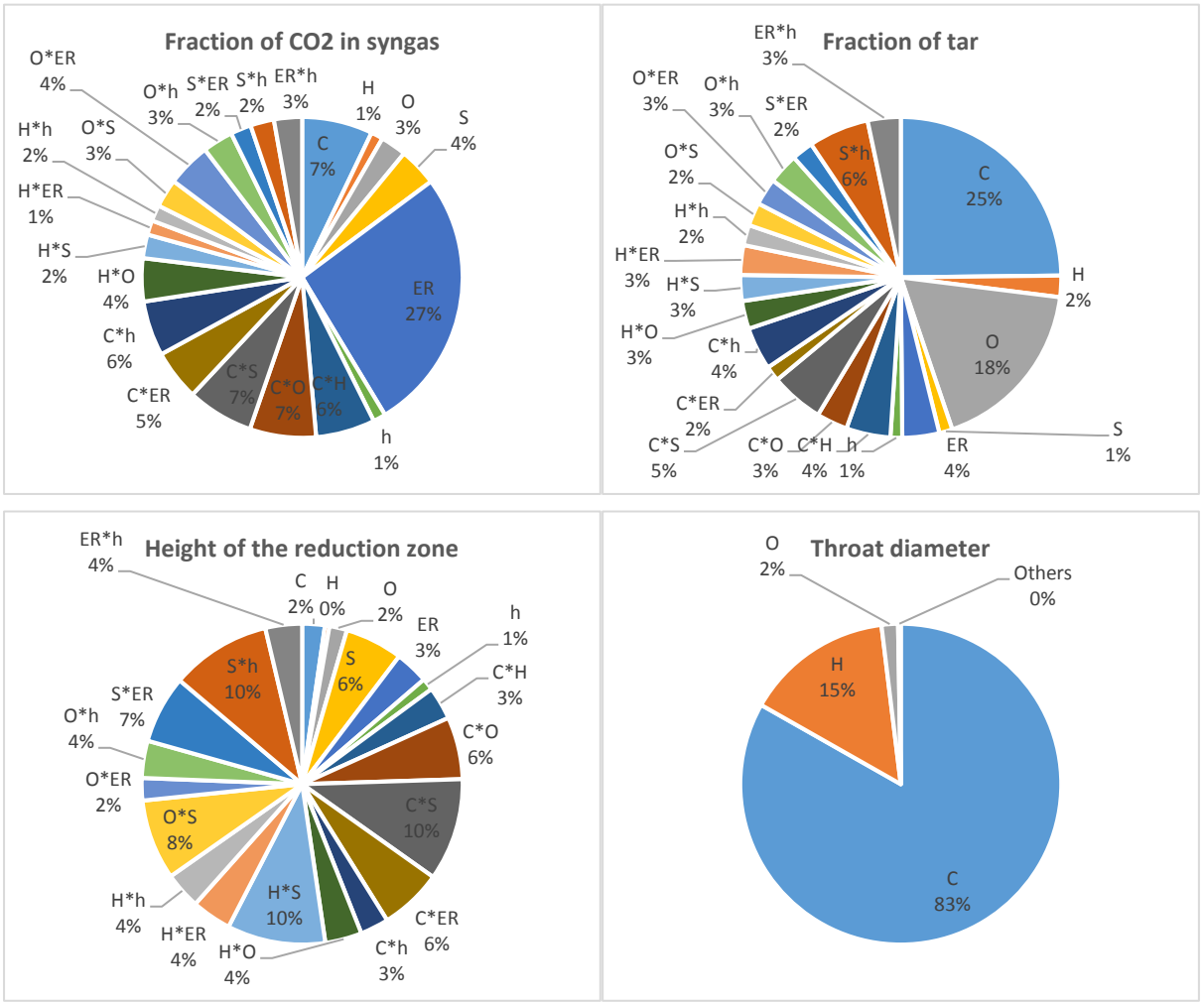
By looking at Figure 6, we can see how key outputs, like the fraction of hydrogen in the syngas and the tar content, are strongly influenced by the temperature of the drying zone, or how the diameter of the pyrolysis zone is predominantly affected by the power output of the gasification unit. These and other conclusions often cannot be directly inferred from the model without the help of advanced statistical analysis algorithms, which is the approach followed and promoted by this work.

Figure 6 Global sensitivities of key outputs to input variables and model parameter for the biomass downdraft gasifier [38]



From Figure 7 we also see how, for example, any uncertainty in the characterisation of the biomass feedstock substantially affects some important outputs and design parameters of the gasifier. In particular, we can see how the carbon content of the feedstock has a strong influence on the throat diameter of the downdraft gasifier. The relatively large number of interconnected terms contributing significantly to the overall uncertainty of the outputs shows that many of the decision variable used by the model are strongly correlated.

Figure 7 Contribution of selected decision variables to key outputs uncertainties for the biomass downdraft gasifier [38]



Sensitivity analysis and uncertainty propagation studies were performed on the model to identify those model parameters, including reactions rate parameters, and decision variables that have the strongest effect on the model outputs. As done for the biomass pyrolysis model developed in *k*inetics, we initially calibrated the model against existing experimental data and compared the results with those of the original model to see if adjustments of the model parameters were required. A comparison between the simulation results from the original and

the calibrated model is presented in Figure 8. A general improvement in the match with the experimental results can be observed when using the calibrated model. Table 3 presents the full set of calibrated parameters used to generate the results shown in Figure 8.

Table 2 Model parameter acronyms [38]

Acronym	**Parameter**
C	Carbon in feedstock
H	Hydrogen in feedstock
O	Oxygen in feedstock
S	Sulphur in feedstock
ER	Equivalence ratio
h	Moisture content
T01	Initial temperature of feedstock
HR1	Heating rate in drying zone
Td	Drying temperature
Ed, Ad	Kinetic parameter for drying process
A1,2, 3, 4	Pre-exponential multipliers for reduction reactions
E1, 2, 3, 4	Activation energy for reduction reactions
T	Reduction zone temperature
Tar1, 2, 3	Multipliers for tar concentration in pyrolysis and oxidation models
char	Multipliers for char concentration in pyrolysis models
PO	Power Output

The calibrated model was then used to optimise the process via MoDS. Major targets considered in this study were the minimization of undesired by-products (e.g. tar), the maximization of the syngas yield and heating value, and the production of specified composition for the syngas (e.g. specific CO/H_2 ratio for implementation of the syngas as chemical feedstock, or maximization of the H_2 ratio in the syngas for hydrogen separation and purification). Optimisation routines used in MoDS include the Hooke and Jeeves direct search solution method, the Metropolis-Hastings algorithm, the truncated Newton method and the COBYLA (Constrained Optimisation by Linear Approximation) algorithm.

Results from a multi-objective optimisation aimed at minimizing the tar content, as well as the fraction of CO_2 and CH_4 (undesired, for example, in H_2 refinery or Fischer-Tropsch processes), is presented in Figure 9. Initial and optimised process conditions and design specifications for the 20 kW downdraft gasifier are reported in Table 4. The optimisation routine (Hook and Jeeves method, for the particular example shown here) was able to

effectively identify a set of optimal conditions and design details leading to a considerable improvement of the syngas quality.

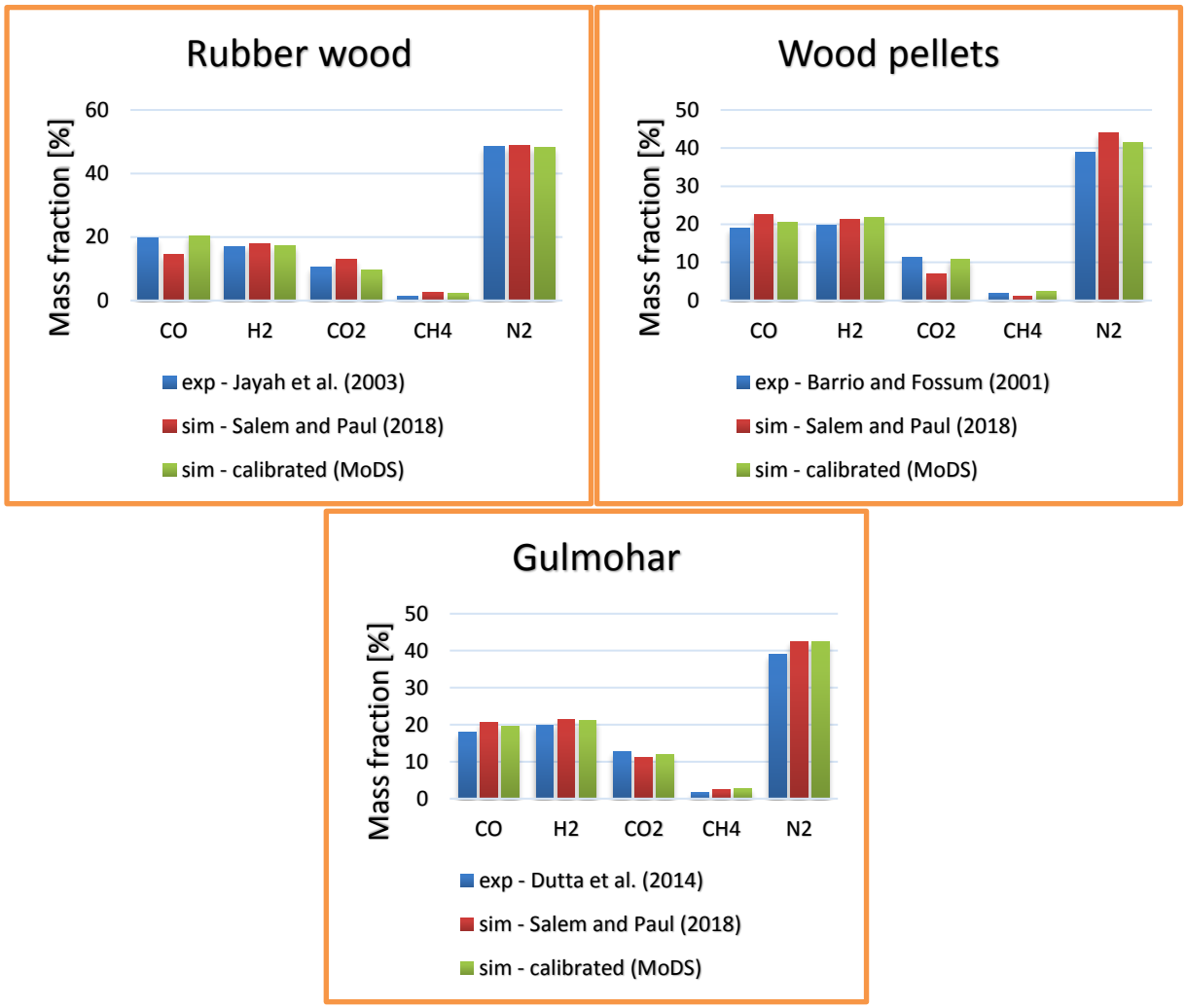


Figure 8 Comparison of syngas composition as simulated by the original model of Salem and Paul [38] and its calibrated version (MoDS) versus experimental results for different feedstocks [48] [49] [47].

Table 3 Calibrated model parameters (Arrhenius coefficients for the chemical kinetic mechanism) [38]

Acronym	Model parameter	Unit	Initial value	Calibrated value
Ed	Activation energy - drying	kJ/mol	88	90.75
Ad	Frequency factor - drying	s^{-1}	5.13E+05	5.24E+10
A1	Frequency factor - reduction (R1)	s^{-1}	36.16	35.715
A2	Frequency factor - reduction (R2)	s^{-1}	1.52E+04	1.51E+04
A3	Frequency factor - reduction (R3)	s^{-1}	4.19E-03	4.24E-03
A4	Frequency factor - reduction (R4)	s^{-1}	7.30E-02	7.42E-02
E1	Activation energy - reduction (R1)	kJ/mol	77.39	77.87
E2	Activation energy - reduction (R2)	kJ/mol	121.62	121.62
E3	Activation energy - reduction (R3)	kJ/mol	19.21	19.12
E4	Activation energy - reduction (R4)	kJ/mol	36.15	35.303

Figure 9 Optimised process outputs (minimisation of tar content and undesired by-product (CO_2 and CH_4) fractions in the syngas) for a 20 kW biomass downdraft gasifier

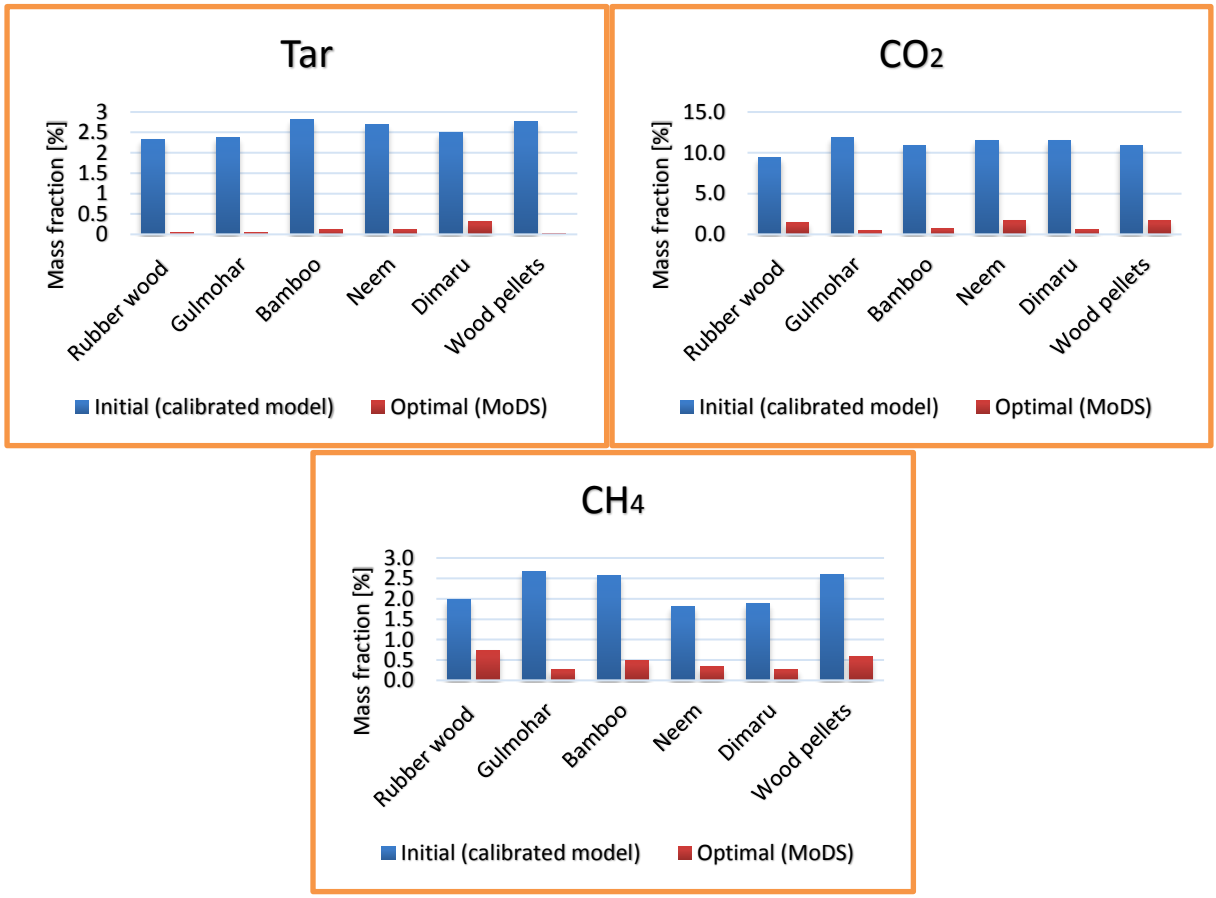


Table 4 Initial and optimal values of operating conditions and design specifications for a 20 kW biomass downdraft gasifier

Parameter	Unit	Initial	Optimal
Equivalence ratio	-	0.3	0.44
Power output	kW	20	22.4
Temperature of the drying zone	K	400	470
Heating rate in drying zone	K/s	20	16.5
Temperature of the pyrolysis zone	K	873	873
Temperature of the reduction zone	K	1350	1139
Height of the drying and pyrolysis zone	cm	57.14	57.14
Height of the oxidation zone	cm	10.09	12.36
Diameter of the pyrolysis zone	cm	20.17	24.71
Throat diameter	cm	5.76	7.06
Area of air injection	cm^2	2.35	3.52
Air injection diameter	cm	16.14	19.77

CONCLUSIONS

Rapid and cost-effective technology development, the design of flexible processes able to efficiently transform different types of biomass, including wastes, into a number of products (e.g. electricity, fuels and chemicals), while simultaneously reducing pollutant emissions and unwanted by-products, can be effectively achieved only through the implementation of integrated virtual engineering techniques. These requirements, coupled with the complexity and uncertainty of the chemistry for biomass pyrolysis and gasification, the variability and unpredictability of biomass feedstock characteristics, and the strong sensitivity to changes in the operating conditions of the process outputs, have proven challenging in the past and have led to a limited exploitation of biomass pyrolysis and gasification, critical technologies in achieving the sustainability of energy supply and industrial decarbonisation governments are currently aiming for worldwide. The implementation of automated calibration and optimisation techniques via direct integration of detailed physico-chemical models with statistical analysis toolkits (MoDS) was used in the current work in the attempt to simultaneously solve the multiple challenges listed above. Process insight was obtained through global sensitivity analysis and uncertainty propagation studies. By using flexible and non-computationally expensive mathematical models, we were able to achieve detailed system understanding, perform robust model tuning, and identify optimal design specifications and operating conditions for the technologies under study in a time frame compatible with those required by real industrial applications. The results obtained from this

study are encouraging and we believe will promote the implementation of similar strategies to a wider range of engineering problems.

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